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NATIONAL INVENTORY OF SOURCES AND EMISSIONS: MERCURY, 1968

W. E. Davis and Associates

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September 1971

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Research Triangle Park, North Carolina 27711

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NATIONAL INVENTORY OF SOURCES AND EMISSIONS: MERCURY - 1968

by

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Contract No. CPA-70-128

EPA Project Officer: C. V. Spangler

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Office of Air Quality Planning and Standards
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Publication No. APTD-1510

PREFACE

This report was prepared by W. E. Davis & Associates pursuant to Contract No. CPA 70-128 with the Environmental Protection Agency, Office of Air Programs.

The inventory of atmospheric emissions has been prepared to provide reliable information regarding the nature, magnitude, and extent of the emissions of mercury in the United States for the year 1968.

Background information concerning the basic characteristics of the mercury industry has been assembled and included. Process descriptions are given, but they are brief, and are limited to the areas that are closely related to existing or potential atmospheric emissions of the pollutant.

Due to the limitation of time and funds allotted for the study, the plan was to personally contact fifteen to twenty percent of the companies in each major emission source group to obtain the desired information. It was known that published data concerning emissions of the pollutant were virtually nonexistent, and numerous contacts with industry during the study ascertained that atmospheric emissions were not a matter of record.

The mercury emissions and emission factors that are presented are based on the summation of information obtained from production and reprocessing companies that handle about thirty percent of the mercury consumed in the United States. Mercury emissions and emission factors are considered to be reasonably accurate.

ACKNOWLEDGEMENTS

This was an industry oriented study and the authors express their appreciation to the many companies and individuals in the mercury industry for their contributions.

We wish to express our gratitude for the assistance of the various societies and associations, and to the many branches of the Federal and State Governments.

Our express thanks to Mr. C. V. Spangler, Project Officer, Office of Air Programs, for his helpful guidance.

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An inventory of atmospheric emissions has been prepared to determine the nature, magnitude, and extent of the emissions of mercury in the United States for the year 1968. The flow of mercury has been traced and charted, indicating that the consumption was 2,866 tons while domestic production of primary and secondary mercury was 2,403 tons. Imports, principally from Spain, Canada, and Mexico totaled 883 tons. Emissions to the atmosphere during the year were 840 tons. About 30% of the emissions resulted from the combustion of coal. Other significant emissions were due to the use of paint, the processing of mercury, and the use of mercury in the electrolytic preparation of chlorine and caustic soda. Emission estimates were based on observations made by personal contact and on information provided by mining, processing and reprocessing companies.

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SUMMARY

The flow of mercury in the United States has been traced and charted for the year 1968. The consumption was 2,866 tons (75,422 flasks) while domestic production of primary and secondary mercury was 2,403 tons (63,200 flasks). Imports, principally from Spain, Canada, and Mexico, totaled 883 tons (23,246 flasks).

Emissions to the atmosphere during the year were 840 tons. About 30 percent of the emissions resulted from the combustion of coal. Other significant emissions were due to the use of paint, the processing of mercury, and the use of mercury in the electrolytic preparation of chlorine and caustic soda.

Emission estimates are based for the greatest part on observations made by personal contact and on information provided by mining, processing, and reprocessing companies. Emissions due to the combustion of coal are based on the only data available, which are relatively incomplete.

EMISSIONS BY SOURCE
1968

Source Category	Source Group	Emissions - Tons		Emissions %
Mining			2.6	0.3
Ore Processing			55.0	6.5
Secondary Production			11.0	1.3
End Product Uses			365.6	43.5
	Paint	216.0		
	Agricultural	19.0		
	Pharmaceuticals	2.6		
	Electrical Apparatus	3.0		
	Electrolytic Chlorine	70.2		
	Instruments	2.6		
	Dental Preparations	1.2		
	General Laboratory Use	51.0		
Other Emission Sources			405.8	48.4
	Coal	255.0		
	Oil	5.0		
	Incineration	10.8		
	Sewage and Sludge	11.0		
	Miscellaneous	124.0		
TOTAL			840.0	100.0

EMISSIONS BY REGIONS

	Number of Plants		Short Tons
	Processing	Chlorine	
Region No. 1	82	3	139
Region No. 2	-	3	256
Region No. 3	3	9	208
Region No. 4	-	8	218
Undistributed			19
TOTAL			840

MAP OF EMISSION REGIONS
SHOWING LOCATIONS OF MERCURY MINES AND CHLOR-ALKALI PLANTS

Figure I

EMISSION FACTORS

The emission factors given herein are believed to be the best currently available. They were determined through a combination of methods consisting of: (1) direct observation of emission data and other related plant processing and engineering data where available; (2) estimations based on information obtained from literature, plant operators, and others knowledgeable in the field; (3) calculations based on experience and personal knowledge of metallurgical processing operations; and (4) specific analytical results (in the case of coal) where available and judged dependable. More reliable data should become available within one or two years after the release of this report.

The basic data used to calculate the emission factors are contained in the files of the Contractor and the Office of Air Programs of the Environmental Protection Agency. Readers of this document are encouraged to submit data to the EPA in confirmation of these factors, or additional data which can be used to further refine the factors in subsequent publications.

A summary of the emission factors is shown below.

Mining	0.012 lb/ton of ore mined
Ore Processing	0.254 lb/ton of ore processed
Secondary Production	40 lb/ton of mercury processed
End Product Uses	
Paint	1,300 lb/ton of contained mercury
Agricultural	1,000 lb/ton of contained mercury
Pharmaceuticals	400 lb/ton of contained mercury
Electrical Apparatus	8 lb/ton of mercury used
Electrolytic Production of Chlorine	0.0585 lb/ton of chlorine produced
Instruments	17 lb/ton of contained mercury
Dental Preparations	20 lb/ton of contained mercury
General Laboratory Use	1,500 lb/ton of total mercury loss
Other Emission Sources	
Coal	1 lb/1,000 tons of coal burned
Oil	0.0000167 lb/bbl of residual oil consumed
Incineration	1.4 lb/1,000 tons of refuse burned

NOTE - All emissions stated in lb refer to lb of mercury.

MINERAL SOURCES OF MERCURY

Mercury is a high density, silver white metal that has uniform volume expansion and is liquid at normal ambient temperatures. It is relatively rare, but it commonly exists in highly concentrated ores found near the earth's surface and is readily attainable. Of twenty-five minerals known to contain mercury, the chief source is cinnabar, red mercuric sulfide (HgS). Important deposits are located in the United States, the Philippines, China, Italy, Mexico, Peru, Spain, Yugoslavia, and the U.S.S.R.

In the United States ores containing mercury are found in Alaska, Arizona, Arkansas, California, Idaho, Nevada, Oregon, Texas, and Washington. During 1968 mercury produced in California and Nevada accounted for ninety percent of the domestic primary production.

MATERIAL FLOW THROUGH THE ECONOMY

The sources and uses of mercury in the United States during 1968 are shown in Figure II. The supply was from three sources: (1) 1,097 tons from primary production; (2) 1,306 tons from secondary production including Government releases; and (3) 883 tons from foreign sources.

Mercury Imports - In 1968 mercury imports into the United States totaled 883 tons (23,246 flasks), approximately 31 percent of the domestic consumption. About 55 percent of the imports were from Spain, 24 percent from Canada, 10 percent from Mexico, 5 percent from Yugoslavia, and 5 percent from Peru [1].

Mercury Exports - Exports and reexports of mercury during 1968 were 289 tons (7,599 flasks). Exports more than doubled as compared with 1967, principally as a result of increased shipments to India and Japan. Foreign trade in mercury compounds was insignificant [1].

Mercury Stocks - Stocks at the beginning of 1968 were

1- Minerals Yearbook; Bureau of Mines; 1968.

18,277 flasks, and at the end of the year the total on hand was 21,484 flasks, an increase of 122 tons (3,207 flasks) 1/.

1- Minerals Yearbook; Bureau of Mines; 1968.

MERCURY

MATERIAL FLOW THROUGH THE ECONOMY - 1968

(Short Tons)

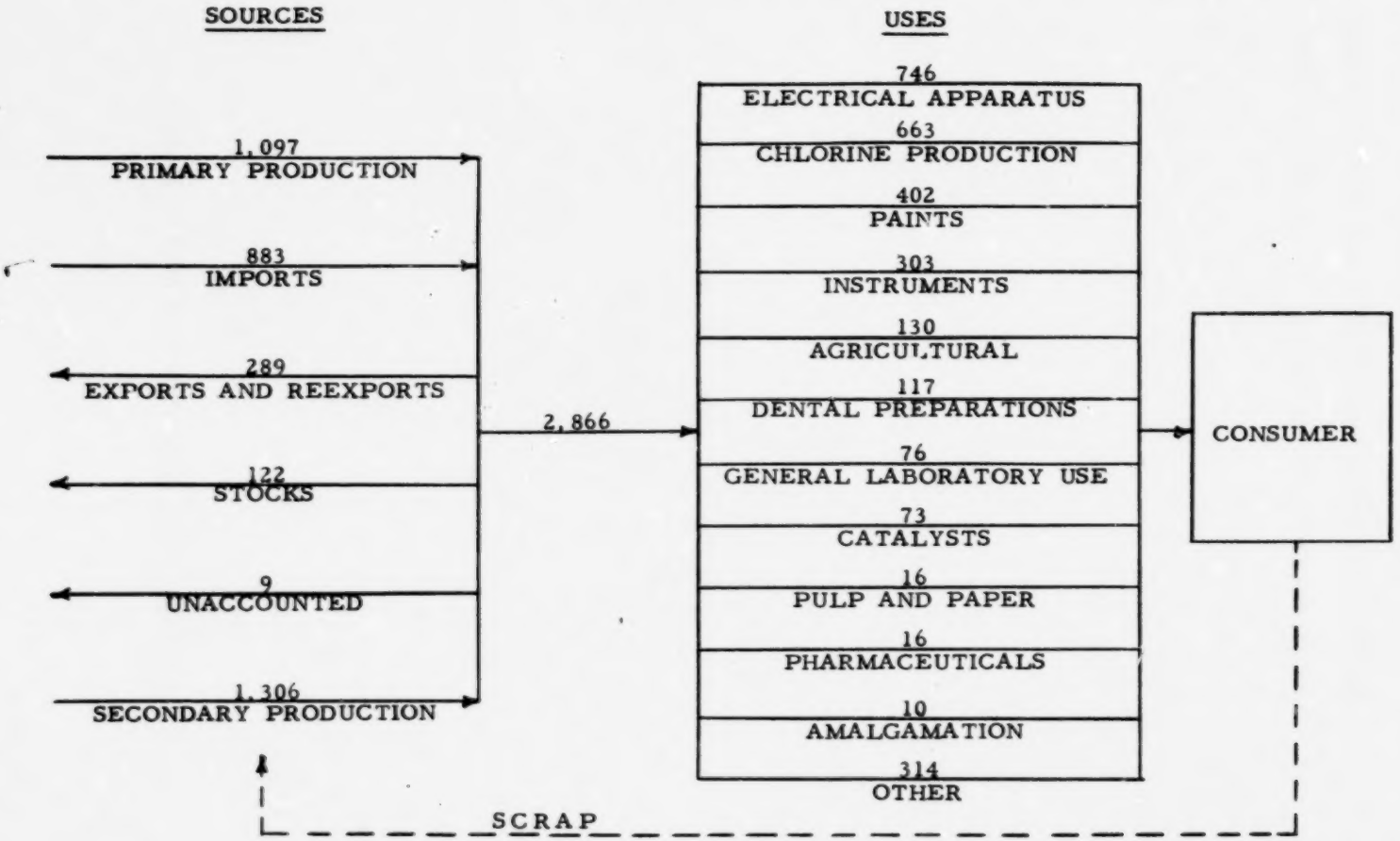

Figure II

USES AND EMISSIONS OF MERCURY

MINING

The mercury consumed in the United States during 1968 totaled 75,422 flasks (76 pounds per flask); however, only 28,874 flasks were produced from domestic ore [1].

The price per flask held above $500.00 throughout the year and many of the mines in operation were producing relatively low quality ores containing only 1.7 to 2.7 pounds of mercury per ton; however, one mine visited during this study reported an average mercury content of 20 pounds per ton of ore. The average for all ore produced was 5.1 pounds of mercury per ton of ore [1]. Since prices remained relatively high, established producers were encouraged to operate mines and plants at near maximum capacities. Of the 87 operations reported for the year, 53 were in California, 17 in Nevada, 6 in Oregon, 3 each in Arizona and Texas, 2 each in Alaska and Idaho, and one in Washington.

In the United States mercury ore is mined by both surface and underground methods. Usually the deposits are small

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1- Minerals Yearbook; Bureau of Mines; 1968.

and irregular; consequently, large-scale mining operations are the exception rather than the rule.

Before mining ore from open-pit deposits near the surface, all barren or low grade material overlying the deposit is removed. The ore is then drilled and broken prior to loading into trucks for transportation to the mill. During open-pit mining operations the mercury emissions to the atmosphere are due to the dust generated during drilling, blasting, and handling the ore, as well as natural vaporization of mercury from the ore deposit.

Even though underground methods of mining mercury ore are not uniform, drilling, blasting, and handling are part of the operations. After the ore has been broken by drilling and blasting it is removed by scrapers, by direct drop to drawpoints, or by mechanical loaders. The ore is either trammed or hoisted to the surface.

Emissions from Mining - Emissions from underground mines are thought to be essentially the same as those from open-pit mines. Concentrations of dust and vapor in the mines must be maintained at a low level in order to protect the miners; therefore, forced ventilation is required. The

systems used are usually rather large and they discharge to the atmosphere.

Accurate data is not available concerning mercury emissions to the atmosphere from sources of mining; however, an estimate has been prepared based on information obtained from mine operators.

Emissions from mining operations in the United States during 1968 totaled 2.6 tons.

ORE PROCESSING

Two general types of recovery are used to produce elemental mercury from cinnabar, the principal ore that is used for the production of mercury.

One system used in the smaller plants consists of a retort into which a batch of crushed cinnabar is dumped, the opening is closed, and heat is applied (1,500 to 1,800 F), and mercury is driven off. The mercury is vaporized by indirect heating and only enough air is admitted to the retort to oxidize the sulfur to sulfur dioxide. The mercury vapor, sulfur dioxide, and any excess oxygen, plus all of the nitrogen, flows to a condenser where the mercury is condensed and the other components are cooled by cooling water or by air. The gases are usually cooled to a temperature ranging from 110 to 140 F. Emissions of mercury into the atmosphere from this system stem from two main sources:

1 - Mercury remaining in the discarded calcine;

2 - Mercury in the inert gas stream discharged from the condenser.

In the retort system the inert gas stream is small and the mercury vapor loss from this source is negligible.

Larger plants usually are equipped with a continuous furnace. The crushed and sized ore flows into the colder end of the furnace which is, in many cases, a rotary kiln type. The kiln is fired at the end opposite the feed. The hot gases contact the ore and heat it to about 1,800 to 2,000 F. The mercury is vaporized by direct heating, the sulfur portion of the cinnabar is oxidized to sulfur dioxide, and the total gas mass flows to the condenser where the mercury vapor is condensed and the noncondensable gases are cooled to approximately 110 F. The exit temperature of the gases is subject to wide variation and depends on the design of the plant. Mercury emissions in this process also stem from two main sources:

1 - Mercury remaining in the discarded calcine;

2 - Mercury in the inert gas stream discharged from the condenser.

This process emits more mercury to the atmosphere at any given temperature of the gas mass than the retort process, because the heating gases in the continuous process are in direct contact with the ore and the gas volume is much larger.

Emissions from Ore Processing - Mercury emissions to

the atmosphere from sources of primary processing are estimated at 55 tons for the year 1968, based on an emission factor of 0.254 pound per ton of ore processed. The emission factor is based on the following information:

(a) Ore processed during 1968 (tons) [1] - 432,772

(b) Condenser exit temperature (F) - 110

$$\frac{432,772 \times 0.254}{2,000} = 55$$

1- Minerals Yearbook; Bureau of Mines; 1968.

MERCURY SECONDARY PRODUCTION

Secondary production of mercury, as reported by the Bureau of Mines, increased about 50 percent during 1968 principally due to releases by the General Services Administration. Reclaim from battery scrap, dental amalgams, and various sludges contributed to the increased production and was a significant part of the domestic supply. Secondary production of mercury during 1968 was 1,306 short tons (34,380 flasks) [1]/ including the GSA releases of 745 tons (19,610 flasks) [2]/. The remaining 561 tons were reclaimed from the dental amalgams, battery scrap, and various sludges.

Emissions from Secondary Production - Based on the Contractor's estimated emission factor of 40 pounds per ton of secondary mercury processed, the atmospheric emissions in the United States during 1968 totaled 11 tons.

1- Minerals Yearbook; Bureau of Mines; 1968.

2- "Trends in Usage of Mercury"; National Materials Advisory Board; NMAB-258; Sept. 1969.

END PRODUCT USES OF MERCURY

The reprocessing of mercury for end product uses is somewhat different than the reprocessing of most metals. In many cases it is used in the elemental form (as a liquid metal) and installed as a working fluid in manufactured products such as electrical switches, thermometers, and rectifiers (Table I). After the manufacturing operations are complete the mercury is "sealed-in" and there is no atmospheric emission problem during everyday use. However, an unknown number of such devices are eventually scrapped and there is an emission from waste disposal and incineration.

During 1968 about 40 percent of the mercury used in the United States was installed in equipment as a working fluid, and 30 percent was used as liquid metal in the production of chlorine and caustic soda. Most of the remaining 30 percent was reprocessed into compounds used in paint, agricultural sprays, pharmaceuticals, catalysts, and other products tabulated in Table II.

The chief applications of the principal compounds are as follows:

Mercuric acetate	- manufacture of organic mercurials.
Mercuric cyanide	- antiseptic in medicine.
Mercurous chloride	- fungicides and insecticides.
Mercuric bichloride	- production of other mercury compounds; seed treatment in agriculture; catalyst in organic reactions; antiseptic in medicine; to intensify negatives in photography.
Mercuric iodide	- analytical reagent; skin streatment in medicine.
Mercuric nitrate	- production of other mercury compounds.
Mercuric oxides	- germicide in medicine; fungicide and pesticide in agriculture; antifouling paints; other mercury compounds; dry cell batteries.
Mercurous sulfate	- constituent of standard cells.
Mercuric sulfate	- catalysts.
Mercuric sulfide	- pigments.

The consumption of mercury in the United States during 1968 has been reported at 2,866 tons (75,422 flasks) 1/.

1- Minerals Yearbook; Bureau of Mines; 1968.

TABLE I

APPLICATIONS WHERE MERCURY
IS USED IN THE ELEMENTAL FORM

Item	Flasks	Short Tons
Electrical Apparatus	19,630	746
Electrolytic Preparation of Chlorine	17,453	663
Industrial and Control Instruments	7,978	303
Dental Preparation	3,079	117
General Laboratory Use	1,989	76
Other	8,275	314
TOTAL	58,404	2,219

TABLE II

APPLICATIONS WHERE
MERCURY COMPOUNDS ARE USED

Item	Flasks	Short Tons
Paint	10,566	402
Agricultural	3,430	130
Catalysts	1,914	73
Pulp and Paper	417	16
Pharmaceutical	424	16
Amalgamation	267	10
TOTAL	17,018	647

Paint - Mercurial compounds are widely used in paint manufacturing, as a toxicant in marine antifouling paints, as a mildew proofing agent in mildew proof paints, and as a paint preservative in latex paints. According to one source of information some manufacturers use mercury in about 95 percent of their products, while others use it in only 10 to 20 percent of the paint they produce. It is used in nearly all latex and alkyd formulas.

The heavier mercurial compounds that are relatively nonvolatile are favored for use in exterior paints because they diffuse to the surface more slowly and protect the paint for a longer period of time. Phenyl mercuric acetate (PMA) and phenyl mercuric dodecyl succinate (PMS) are two of the phenyl mercury compounds most commonly used as fungicides in paints; however, in recent years phenyl mercuric dimethyldithiocarbamate (PMDDC) has also been used for this purpose. It has been demonstrated that the retention of PMS in emulsion paint films is slightly better than the retention of PMA, but that PMDDC is retained much better than either of the others [1]. The high retention of PMDDC

1- Taylor, G. C., Tickle, W. and Dwyer, A.; "Radiometric Studies of Mercury Loss from Fungicidal Paints - II"; J. Appl. Chem.; 19; Jan. 1969.

is probably due to its low saturation vapor pressure (Table III) and its low solubility in water.

The report of a study conducted for the National Paint, Varnish and Lacquer Association during September, 1964 [1] compares the results of tests made before, during, and after painting the walls and ceiling of a room with latex paint containing 0.02 percent mercury. Concentrations reached a value of 0.17 mg/m^3 in about 90 minutes and remained at that level until the painting was completed. After 24 hours mercury concentrations had fallen to 0.01 mg/m^3. It is apparent that interior paints containing mercury compounds emit mercury into the atmosphere and the rate of emission is rapid while the paint is wet.

Concentrations of mercury in the paints usually vary from 0.02 to 2.5 percent depending on the use for which they are intended, and the rate of the emission is directly related to the concentration in the paint [2]. Mildew resistant paints generally contain the most mercury and usually provide mildew protection for a period of two or three years. In other

---

1- Goldwater, L. J. and Jacobs, M. B.; "Mercury Exposure from the Use of a Mercury-Bearing Paint"; Unpublished report; Nov. 9, 1964.

2- Private communication with paint manufacturer.

words, most of the mercury compound in the paint has diffused to the surface within two or three years and has been emitted to the atmosphere or washed down into the soil and water.

During manufacturing operations the mercury compounds are added to the paint during final stages of mixing; the entire operation is carried out in closed equipment. Paint manufacturers report that mercury emissions to the atmosphere are exceedingly small.

During 1968, 14 percent of the mercury consumed in the United States was used in paints. During the year, 402 tons (10,566 flasks) were used for that purpose [1].

Emissions from Manufacture of Paint: Based on the above information, mercury emissions to the atmosphere resulting from the manufacture of paint during 1968 are estimated by the Contractor at one ton. The emission factor is 5 pounds per ton of mercury used in the manufacture of paint.

Emissions from Use of Paint: According to the information obtained from paint manufacturers and the results of tests,

---

1- Minerals Yearbook; Bureau of Mines; 1968.

it is apparent that there are substantial mercury emissions to the atmosphere due to the use of paint. It is estimated that 65 percent of the mercury used in paints is emitted to the atmosphere within two or three years after the paint is applied.

Based on an annual use of mercury in paint averaging 331 tons (Table IV), the mercury emissions to the atmosphere during 1968 due to the use of paint are estimated by the Contractor at 215 tons.

Agricultural - During 1968 the use of mercury for agricultural purposes totaled 130 tons (3,430 flasks) [1]. It was used in the form of organomercurial compounds in fungicides and bactericides for control of diseases of fruits, vegetables, and grains. The primary uses were an estimated 54 tons for seed treatment of cereal grains [2], and 38 tons for preparation of sprays. There is no substitute currently available that combines the broad spectrum of control achieved by mercurial compounds.

1- Minerals Yearbook; Bureau of Mines; 1968.

2- "Trends in Usage of Mercury"; National Materials Advisory Board; NMAB-258; Sept. 1969.

TABLE III

VAPOR PRESSURE OF MERCURY COMPOUNDS

	°C	Vapor Pressure mm Hg
Mercury (metal)	36	4.47×10^{-3}
Dimethyl Mercuric Mercury	94	760
Methyl Mercury Chloride	35	32×10^{-3}
Phenyl Mercuric Acetate	35	5×10^{-6}
Phenyl Mercuric Dimethyldithiocarbamate	35	0.8×10^{-6}

TABLE IV

MERCURY USED IN PAINT

Year	Flasks 1/
1966	8,420
1967	7,178
1968	10,566
TOTAL	26,164
Annual Average	8,721 (331 tons)

1- Minerals Yearbook; Bureau of Mines; 1968.

From the standpoint of atmospheric emissions, the sprays containing mercurial salts and compounds are partially washed from the foilage into the soil. However, the part that remains on the plant is exposed and emissions to the atmosphere result. The U. S. Department of Agriculture and others recognize there are mercury emissions to the atmosphere due to spraying, but data are not available regarding drift and off-target problems.

The methylation of mercury by enzymatic and non-enzymatic processes has been reported by Wood [1] and Jensen [2] but no statement has been made regarding the escape of methyl mercury from water into the air. It has been reported that mercury compounds in the soil are converted to metallic mercury [3].

Emissions from the Use of Agricultural Sprays: Based upon the information obtained during this study it is assumed that

---

1- Wood, J. M., Kennedy, F. S. and Rosen, C. G.; "Synthesis of Methyl-mercury Compounds by Extracts of a Methanogenic Bacterium"; Nature; 220; Oct. 12, 1968.

2- Jensen, S. and Jernelov, A.; "Biological Methylation of Mercury in Aquatic Organisms"; Nature; 223; Aug. 16, 1969.

3- Booer, J. R.; "The Behavior of Mercury Compounds in Soil"; Annals of Applied Biology; 31; pp. 340-359; Nov. 1944.

50 percent of the mercury used in the preparation of sprays will become an atmospheric emission. Therefore, the Contractor's estimate is that 19 tons of mercury were emitted to the atmosphere during 1968. The emission factor is 1,000 pounds per ton of mercury contained in the spray materials.

It is assumed that mercury used as a seed treatment or a soil conditioner will be introduced directly into the soil and that atmospheric emissions will be negligible.

Catalysts - Organic mercurial salts are used in the production of urethane elastomers, vinyl chloride monomers, sulfonated anthraquinone products, and for numerous miscellaneous purposes (Table V). One of the outstanding new uses in 1968 was the incorporation of organic mercury catalysts in urethane resins molded into automobile bumpers on one series of an intermediate priced car. About 3 parts of the commercial mercury catalyst are employed in 100 parts of resin. Active mercury content in the catalyst is in the order of 5 percent [1]/..

1- "Trends in Usage of Mercury"; National Materials Advisory Board; NMAB-258; Sept. 1969.

TABLE V

USE OF MERCURY AS A CATALYST

Item	Flasks	Short Tons
Urethane	800	30
Vinyl Chloride Monomer	500	19
Anthraquinone Derivatives	175	7
Miscellaneous	439	17
TOTAL [1]/	1,914	73

1- Minerals Yearbook; Bureau of Mines; 1968.

The chlorides, oxides, sulfates, acetates, and phosphates are used in catalysts in various ways. The processes for making these materials are relatively simple one-step reactions usually carried out in batch operations. Mercury molecules are very heavy and if covered by a liquid, diffusion into the atmosphere is very slight during the reaction period.

Emissions from the Manufacture of Catalysts: Losses from such compounds as mercuric sulfate, mercuric oxide, mercuric chloride, and mercuric phosphate during reaction and packaging is considered to be negligible.

Pulp and Paper - The use of mercury compounds in the pulp and paper industry has declined substantially since 1963. Decreased use is also anticipated for the future; however, the high degree of effectiveness of mercury compounds and their ability to prevent the formation of slime during paper manufacture seem to indicate there will be a market in this area.

In the United States 16 tons (417 flasks) of mercury were used in pulp and paper manufacture during 1968 [1].

1- Minerals Yearbook; Bureau of Mines; 1968.

Emissions from Manufacturing: The special mercury compounds used in the pulp and paper industry are produced in batch operations in relatively small amounts, and most of the materials used have a low vapor pressure. Losses of mercury to the atmosphere due to the manufacture of slimicides is considered to be negligible.

Emissions from Use: During paper making mercurial biocides are added to slurries of cellulose fibers to reduce the growth of slimes in the slurry which cause difficulty in the paper-making machine. During the process most of the mercury compound that is used is removed from the slurry; however, a small part adheres to the cellulose fibers.

Mercury emitted to the atmosphere due to the use of mercurials in paper making during 1968 is considered to be negligible.

Pharmaceuticals - It has been reported that 16 tons (424 flasks) of mercury were used in various pharmaceutical applications during 1968 (Table VI) 1/. It is used in the form of ammoniated mercury, yellow mercuric oxide, or prime virgin mercury in a variety of therapeutic and cosmetic skin creams. One of the purposes is to "fade blemishes, brown

---

1- Minerals Yearbook; Bureau of Mines; 1968.

TABLE VI

MERCURY CONSUMPTION IN PHARMACEUTICALS

Item	Flasks	Short Tons
Diuretics	70	3
Antiseptics	180	7
Skin Preparations	140	5
Preservatives	34	1
TOTAL	424	16

spots, and dull dark areas". It is thought that the compounds used in this manner are free to exert their full vapor pressure and will evaporate from the skin or affected area. Mercury compounds have been used as diuretics for many years, but this use is now declining. In cosmetics and soaps those used as preservatives include phenyl mercuric acetate, phenyl mercuric borate, phenyl mercuric benzoate, and phenyl mercuric nitrate.

Many diuretics, antiseptics, skin preparations, and preservatives are complicated organomercurial compounds that require several reaction steps during processing. There are some, however, that are relatively simple and require only one step. Regardless of the number of processing steps, the operations are normally carried out in enclosed equipment or are vented through condensers.

During use of the various products the mercury is fully expended and enters the environment as a pollutant, partially as an emission to the atmosphere. Mercury contained in diuretics is emitted to water and very little, if any, to the atmosphere. Mercurials in antiseptics are emitted partially to water and partially into the atmosphere. Mercurials

present in skin preparations and ointments are not a portion of the solution and they vaporize to some extent, becoming an atmospheric pollutant. Mercury added to soaps and face creams as a preservative will also evaporate into the atmosphere.

Emissions from Manufacturing: Manufacturers' records of mercury emissions are not available; however, the Contractor's estimate has been prepared assuming the loss to be 10 pounds per ton of mercury processed. The estimate indicates mercury emissions to the atmosphere are negligible.

Emissions Resulting from Use: Since information is not available regarding atmospheric emissions of mercury due to the use of pharmaceuticals, it is assumed for this report that emissions due to the use of diuretics are negligible. For antiseptics, skin preparations, and preservatives, the assumed emission factor is 400 pounds per ton of contained mercury. Based on these assumptions mercury atmospheric emissions for 1968 were 2.6 tons.

Amalgamation - Most metals can be amalgamated with mercury, the one important exception being iron. In the early days mercury was used in mining to recover free gold and

silver from placer and lode ores. Potassium, sodium, and zinc amalgams with mercury are used as reducing agents. As an example, sodium amalgam has been used in the production of tetraethyl lead.

Another amalgam metallurgy application is the recovery of zinc from drosses. Mercury forms the moving cathode in an electrolytic process and when extracted from the cell, it carries a small amount of zinc.

During 1968 mercury used in amalgamation totaled 10 tons (267 flasks) [1].

Emissions Resulting from Amalgamation Operations: Most uses appear to fall into two categories: (a) use of an amalgam of mercury in chemical manufacturing operations, and (b) use of mercury in electrometallurgy.

For the processing which is electrolytic in nature there is little, if any, atmospheric emission. For other processing, emissions are estimated by the Contractor at 10 pounds per ton of mercury processed and are considered negligible.

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1. Minerals Yearbook; Bureau of Mines; 1968.

Electrical Apparatus - During the year 1968 an estimated 746 tons (19,630 flasks) were consumed in this rather important use for mercury [1]. The largest single use in this category was in batteries. Mercury is used in the mercury cell and also in the alkaline energy cell. It is used in the form of mercuric oxide mixed with graphite and as a powdered zinc-mercury alloy.

It has been estimated that 13,000 flasks of mercury were used in battery manufacture in 1968; 1,200 flasks to produce fluorescent and high intensity arc discharge lamps, and about 500 flasks in the manufacture of power rectifiers [2].

The use of mercury in electrical apparatus is principally a handling operation in which small amounts of mercury are installed in equipment such as lamps, batteries, and power tubes. Loss of mercury during manufacturing is primarily a function of the factory room conditions and the ventilation system. Data presented by Biram [3] indicate a mercury

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1- Minerals Yearbook; Bureau of Mines; 1968.

2- "Trends in Usage of Mercury"; National Materials Advisory Board; NMAB-258; Sept. 1969.

3- Biram, J. G. S.; "Some Aspects of Handling Mercury"; Vacuum; 5; pp. 77-92; Oct. 1955.

handling loss of 4 percent per year. It is assumed that 10 percent of the handling loss is the atmospheric emission.

Emissions from Manufacturing: Based on the above, mercury emissions to the atmosphere due to the manufacture of electrical apparatus during 1968 totaled 3 tons.

Emissions Resulting from Use: Mercury emissions that occur due to the use of electrical apparatus containing mercury are principally losses associated with breakage. Such losses are unaccounted for in this report due to lack of reliable information. Emissions from industrial plant fires, building fires, and other such mishaps could be substantial. Emissions from incinerators and other solid waste disposal facilities are believed to result principally from the disposal of mercury vapor type lamps.

Electrolytic Preparation of Chlorine - Two types of electrolytic cells are commonly used for the production of chlorine and caustic soda. The most widely used is the diaphragm cell. As the name suggests, it contains a separating diaphragm through which the chlorine ions must pass on their way to a carbon anode where the chlorine is released.

The other cell, called the mercury cell, has gained acceptance during the past twoenty years and at present accounts for about 28 percent of the chlorine production. The cell produces chlorine and a 50 percent solution of caustic soda (directly from the cell) that is relatively free of salt and iron. This caustic is preferred by the rayon industry.

The mercury cell is a horizontal trough, the bottom of which is 30 to 60 inches wide and some 15 to 20 feet long, sloped gradually from inlet to outlet. Mercury flows over this flat surface in a very thin stream and purified brine flows on top of the stream of mercury. DC electric power is applied to the bottom section of the trough and passes to the mercury which acts as the cathode in the system. Sodium ions flow to the cathode and sodium is released at the mercury cathode where it is immediately amalgamated by the mercury. This flows to the end of the cell, out through a seal, and to the denuder which separates the mercury and caustic as the amalgam reacts with water.

The carbon anode is supported above and is in contact with the brine. The anode receives power and has a positive charge; as chlorine ions flow to it, chlorine gas is released

from the brine. Chlorine gas flows from the cell and into the chlorine header.

Distilled water is added to the denuder into which the sodium mercury amalgam flows. Water reacts with the sodium of the amalgam, returning the mercury to its original state and producing a solution (approximately 50 percent) of sodium hydroxide as well as a stream of relatively pure hydrogen, both contaminated with mercury.

The streams of mercury and brine are thin in order to reduce resistance to the flow of current. They must be clean and free of any slimes containing solid material. Solid matter in a thin stream of either mercury or brine will produce a discontinuous film.

The mercury cell operation is quite sensitive to a clean mercury stream, a clean brine stream, and a clean cell. This means regular disassembly, removal of all brine and mercury, and thorough cleaning of the cell. Some mercury is lost in this operation and in the slimes which are removed.

The cells operate at atmospheric pressure, but it is possible that small amounts of mercury vapor may escape from the

cell. Also, mercury "spills" will vaporize from the floor of the cell room. Air change in the cell room is required to hold the concentration of mercury within satisfactory limits (.1 mg/m^3).

Of the 8.4 million tons of chlorine produced in the United States in 1968 approximately 28.5 percent, or 2.4 million tons, was produced in mercury cells [1]. The installed capacity of mercury cells is about 6,900 tons of chlorine per day and mercury inventory is about 90,000 pounds per 100 tons per day of chlorine production; therefore, the active inventory of mercury in the industry is approximately 6.2 million pounds of mercury. Biram reports an inventory loss of 4 percent per annum due to handling mercury [2]. This experience indicates a loss of 0.10 pound of mercury per ton of chlorine produced; however, industrial experience indicates the actual loss is about 0.20 pound of mercury per ton of chlorine produced.

Losses of mercury in the hydrogen stream vary substantially

1- Private communication with the Chlorine Institute.

2- Biram, J. G. S.; "Some Aspects of Handling Mercury"; Vacuum; 5; pp. 77-92; Oct. 1955.

from one plant to another. The hydrogen generated and discharged from the process is handled in different ways. At some locations the hydrogen is cooled and discharged to the atmosphere; at others it is compressed and used as fuel; at still other plants it is compressed and used in the manufacture of ammonia and other hydrogenating or reducing operations. Regardless of how the hydrogen is used, it carries some mercury and there is a mercury loss.

An average operating temperature for the denuder is 180 F. Hydrogen leaving the denuder saturated with mercury will carry a large quantity of mercury vapor (Table VII). Cooling to 90 F can be accomplished and at this temperature the hydrogen will carry 0.028 pound of mercury per ton of chlorine produced. If the hydrogen is compressed to 25 psig, cooled to 90 F, and water is separated from it before it is burned, the estimated loss is 0.01 pound of mercury per ton of chlorine produced.

At some plants the hydrogen is compressed to 400 pounds, cooled to 90 F, and contacted with carbon. This hydrogen is used in making chemicals. The loss of mercury is estimated to be 0.001 pound per ton of chlorine for this higher pressure use of hydrogen.

TABLE VII

MERCURY LOSSES IN HYDROGEN

°F	Pressure mm Hg	Lb/T Cl_2
68	.001201	.015
86	.002777	.021
90	.00374	.028
104	.006079	.046
122	.01267	.0969

In this study it is est. nated that:

(1) 40 percent of the hydrogen byproduct is cooled to 90 F and burned;

(2) 30 percent is compressed to 25 F, cooled to 90 F and burned;

(3) 30 percent is compressed to 400 psig, cooled to 90 F, contacted with carbon, and then used in other chemical operations.

Based on the preceeding conditions, mercury losses are estimated as shown in Table VIII.

TABLE VIII

MERCURY EMISSIONS
ASSOCIATED WITH BYPRODUCT HYDROGEN

Condition	Tons/Year
(1)	13.4
(2)	3.6
(3)	0.4

The estimated loss totals 17.4 tons for the year 1968, or an average loss to the atmosphere in the hydrogen stream of .01454 pound of mercury per ton of chlorine produced.

European practice [1] indicates a loss of 3 percent of the total loss of mercury in the hydrogen, or 0.015 pound per ton. Swedish experience at one plant in 1967 and 1969 is shown in Table IX [2].

TABLE IX

MERCURY LOSSES PER TON OF CHLORINE

	1967 One Plant	1969 One Plant
Loss to water	30-40 g	0.55 g
Loss in hydrogen	5-10 g	0.40 g
Loss to ventilation	15-25 g	1.00 g
Loss to caustic	1-10 g	0.80 g

Emissions from Chlor-Alkali Plants: From calculations made and literature cited, the losses of mercury to the atmosphere are estimated for the year 1968 as shown in Table X.

1- Chlor-Alkali Report; NAPCA; 1970; Study in progress; (unpublished).

2- Private communication with the Chlorine Institute.

TABLE X

MERCURY EMITTED TO ATMOSPHERE

	Avg. lb/ton Chlorine	Tons Mercury
Loss in hydrogen	0.01454	17.4
Loss in ventilation	0.0440	52.8
TOTAL LOSS		70.2

The average loss of mercury to the atmosphere during 1968 is estimated to be 0.0585 pound of mercury per ton of chlorine produced.

During 1968 the chlorine industry used 663 tons (17,453 flasks) of mercury [1] to offset losses that occurred during the manufacturing process.

Industrial and Control Instruments - During 1968 an estimated 303 tons (7,978 flasks) of mercury were used in industrial and control instruments [1] such as barometers, thermometers, flow meters, pressure-sensing devices, switches, and relays. As much as 8 pounds of mercury may be used to equip a flow meter for operation and actual

1- Minerals Yearbook; Bureau of Mines; 1968.

filling of the meter takes place at the location where the instrument is to be placed in service. Thermometers, switches, relays, and most other instruments use considerably less mercury and they are filled at the manufacturing plant.

In the past flow control and metering instruments have been designed using mercury to measure differential pressure and rather large quantities of mercury have been required for this purpose. In such instruments the mercury is in direct contact with the flowing fluid as it flows through an orifice or flow nozzle where the differential pressure is measured by means of pressure taps located on each side of the orifice.

At present industry is eliminating instruments of the type described above and are manufacturing others using pressure transmitting cells that do not require mercury. These newer instruments can be used in the food processing industry as well as for other industrial purposes.

Switches and relays, thermometers, and thermal systems that contain mercury are enclosed devices in which the mercury is "sealed-in". Since glass is used to a great extent, accidental b eakage will result in minor emissions to the atmosphere.

Flow meters and other controls using mercury are, for the most part, installed in industrial plants and the value of the contained mercury is appreciated. Normally the metal will be recovered, cleaned, and reused. It is recognized, however, that during maintenance some mercury will be lost and part will be an atmospheric emission.

Emissions Resulting from Manufacturing: From information obtained through personal contact with manufacturers of instruments and controls, estimates have been made of the quantity of mercury used for various classes of instruments; then, estimates have been made for mercury emissions to the atmosphere. These estimates are shown in Table XI.

TABLE XI

MERCURY CONSUMPTION AND EMISSIONS
INSTRUMENTS AND CONTROLS INDUSTRY

Item	Annual Consumption		Emissions
	Flasks	Tons	Tons
Switches and Relays	2,500	95	0.1
Thermometers	1,000	38	0.04
Thermal Systems	2,000	76	0.08
Flow Measurement	2,478	94	2.4
TOTAL	7,978	303	2.62

Dental Preparations - Mercury is used extensively in dental work as an amalgam which is made up of approximately 50 percent mercury and 50 percent silver and tin. The amalgam for filling cavities in teeth is relatively inert, is easily applied by the dentist, has good compressive strength, has good abrasion resistance, and is relatively permanent. However, in one respect the common "silver filling" is unsatisfactory as a filling material. It is a good conductor of heat and the tooth is subject to thermal shock. Substitutes, such as silicate zinc phosphate cements and acrylic or epoxy resins, have been used but have not yet replaced the mercury amalgam.

The loss of mercury from amalgam fillings has been studied extensively [1] and there is no evidence of change in the mercury content of fillings installed in a person's mouth. It is thought that the amalgam is not well ventilated and is continually covered with water. These two factors minimize the vaporization of mercury from the installed amalgam. The amalgam does not dissolve in body fluids.

1- "Physical Properties of Dental Materials"; National Bureau of Standards; Circular 433; Feb. 6, 1942.

Literature states that gaseous mercury atoms are very heavy and tend to hold over liquid source unless strongly ventilated. Copplestone [1] recommends use of a mixture of calcium oxide, sulfur, and water to suppress vaporization of mercury. Hair spray is also recommended.

The packaging of the materials making up an amalgam has been improved, thereby reducing losses in dental offices. It is recommended that scrap be stored in air-tight containers to limit vapor escape.

Emissions Resulting from Preparation of Dental Amalgams: Under the above conditions, the handling loss should be in the order of 4 percent [2] and emissions to the atmosphere of one percent may be expected.

Mercury used in dental work in the United States in 1968 is estimated at 117 tons (3,079 flasks) [3].

1- Copplestone, J. F. and McArthur, D. A.: "Vaporization of Mercury Spillage", Archives of Environmental Health; 13; p. 675; 1966.

2- Biram, J. G. S.: "Some Aspects of Handling Mercury"; Vacuum; 5; pp. 77-92; Oct. 1955.

3- Minerals Yearbook; Bureau of Mines; 1968.

During 1968 mercury emissions to the atmosphere due to the use of dental preparations totaled 1.2 tons.

General Laboratory Use - Mercury is a common working material in nearly all laboratories of the chemical, physical, and biological sciences. It is used principally to confine gases and is desireable because it does not react with or dissolve in them to any appreciable extent.

The consumption of mercury in laboratories is principally due to spillage and the fact that complete recovery from a spill is impossible. Annual handling losses have been reported at 4 percent of supply [1], but in some college and university laboratories they are known to be much higher, averaging as much as 13 percent of base supply [2]. This is undoubtedly due to inexperienced people performing the experimental work.

Emissions Resulting from Laboratory Use: During 1968 general laboratory use of mercury totaled 76 tons (1,989

1- Biram, J. G. S.; "Some Aspects of Handling Mercury"; Vacuum; 5; pp. 77-92; Oct. 1955.

2- Private communication.

flasks) [1]. In this report it is assumed that 8 tons were used in establishing new laboratories and 51 tons of the remaining 68 tons were atmospheric emissions.

1- Minerals Yearbook; Bureau of Mines; 1968.

OTHER SOURCES OF MERCURY EMISSIONS

COAL

A search has been conducted for information related to the atmospheric emissions of mercury due to the combustion of coal; a limited quantity of recent data regarding the mercury content of coal has been located.

The Illinois State Geological Society commenced testing samples of coal during the latter part of 1970 to determine mercury content. Fifty-five samples of Illinois coal and 11 samples of coal from other states were analyzed for mercury by the neutron activation method. The coal from Illinois was untreated (raw coal) and was from 10 different coal seams currently being mined. The range in mercury content was from 0.04 ppm to 0.49 ppm, and the mean was 0.18 ppm. The mercury in the 11 samples of coal from Arizona, Colorado, Montana, Ohio, Pennsylvania, and Utah ranged from 0.02 ppm to 0.28 ppm [1].

During May, 1971 several coal samples were analyzed for

1- Ruch, R. R., Gluskoter, H. J. and Kennedy, E. J.; "Mercury Content of Illinois Coals"; Illinois State Geological Survey; Environmental Geology Notes No. 43; 1.

the Environmental Protection Agency, Office of Air Programs, using the neutron activation method. The average mercury content for 12 samples of coal from various parts of the United States was 0.81 ppm.

Considering the values reported in the literature, an arbitrary but reasonable average has been selected to estimate emissions for this report. The average mercury content of coal used in the United States during 1968 is assumed to be 0.50 ppm. This is about seven times the geometric mean concentration of mercury in the soil of the United States, which is reported to be 71 ppb [1].

Based on 508,990,000 tons of bituminous and anthracite coal consumed [2], an average mercury content of 0.50 ppm, and all mercury emitted to the atmosphere, the mercury emissions in the United States during 1968 due to the combustion of coal totaled 255 tons.

1- Shacklette, H. T., Boerngen, J.G. and Turner, R. L.; "Mercury in the Environment - Surficial Materials of the Conterminous United States". U. S. Geological Survey Circular 644; 1971.

2- Minerals Yearbook; Bureau of Mines; 1968.

OIL

Until recently data regarding the mercury content of crude and residual oils used in the United States was virtually nonexistent. At the beginning of this study only one electric utility company was able to furnish an analysis that included the mercury content of fuel oil. That company reported the mercury content of oil used during 1968 as 0.15 to 0.60 ppm [1].

Since January, 1971 forty-seven samples of imported residual oil have been analyzed and the average mercury content has been reported as 0.05 ppm. The various samples ranged from a trace to 0.3 ppm [1].

The residual fuel oil used in the United States during 1968, exclusive of use in vessels, was 581.9 million barrels. This oil containing mercury at an estimated 0.05 ppm (average) was used by industrials, electric utility companies, railroads, oil companies, and the military, as well as for heating (Table XII).

1- Private communication.

TABLE XII

SHIPMENTS OF RESIDUAL FUEL OIL IN THE UNITED STATES - 1968

Use		Million Barrels
Heating		174.3
Industrial		175.0
Electric Utilities		185.0
Military and Other		47.6
TOTAL		581.9
States		
Northeast		336.6
New York	116.4	
Massachusetts	67.8	
New Jersey	62.6	
Pennsylvania	42.4	
Connecticut	28.1	
Other	19.3	
South		98.9
Florida	38.6	
Virginia	12.0	
Other	48.3	
Pacific-Mountain		87.3
California	51.8	
Washington	10.0	
Other	25.5	
North Central		59.1
Illinois	24.1	
Indiana	11.4	
Other	23.6	
TOTAL		581.9

"Shipments of Fuel Oil & Kerosine in 1968"; Mineral Industry Surveys; U. S. Dept. of the Interior; Bureau of Mines; Sept. 17, 1969.

TABLE XIII

RESIDUAL FUEL OIL DATA

Residual Oil Burned - 1968 (bbls)	581,900,000
Pounds per Barrel	340
Mercury Content of Oil (ppm)	0.05

Based on the data in Table XIII, the mercury emissions to the atmosphere due to the combustion of residual oil totaled 5 tons during 1968.

INCINERATION

During December, 1970 and January, 1971 a survey was conducted by the State of Illinois Institute for Environmental Quality to determine mercury emissions from power plants, municipal incinerators, and industrial sources. The equipment used was a Barringer Airborne Mercury Spectrometer which was mounted in a helicopter. In general, the measurements of maximum mercury concentration were taken when the helicopter was hovering between 50 and 400 meters downwind from the emission source.

All values reported were based on a very limited number of observations. Data obtained at the municipal incinerator in Chicago, Illinois (Lake Calumet) showed a maximum mercury concentration of 4,450 ng/m^3 which indicated a 0.7 ppm mercury concentration in the refuse that was incinerated [1].

The number of municipal-size incinerators in operation in the United States in 1966 totaled 254; the average capacity

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1- Private communication with William M. Vaughn and Steven B. Fuller; Committee for Environmental Information; Washington University; St. Louis, Missouri.

was 300 tons per day [1]. The total annual capacity was 28 million tons; however, many incinerators were operated at less than capacity. Altogether, 190 million tons of solid waste per year or 5.3 pounds per person per day are collected; approximately 8 percent (15.4 million tons per year) is burned in municipal incinerators [1].

During 1968 mercury emissions to the atmosphere from municipal incinerators was an estimated 10.8 tons.

$$\frac{15,400,000 \times 0.7}{1,000,000} = 10.8$$

1- "Control Techniques for Particulate Air Pollutants"; National Air Pollution Control Administration Publication No. AP-51; Jan. 1969.

SEWAGE AND SLUDGE

A recent report concerning the burning of sewage and sludge indicates that the burning rate in the United States is about 2,000 tons per day, and the mercury content ranges up to 30 ppm [1]/.

Based on the best current estimate, the average mercury content of sewage and sludge is 15 ppm [1]/. On this basis the atmospheric emissions of mercury resulting from the burning of sewage and sludge totaled 11 tons for the year 1968.

1- Private communication from the Federal Water Pollution Control Authority. (Investigations being conducted during 1971; therefore, data are subject to revision.)

MISCELLANEOUS EMISSIONS

Of the 2,866 tons (75,422 flasks) of mercury consumed in the United States during 1968 about 1,627 tons were used for agriculture, paint, general laboratory use, pulp and paper manufacture, pharmaceuticals, electrolytic chlorine plants, and other miscellaneous purposes. It has been considered that this mercury is lost during use or has been reclaimed as a part of the secondary mercury recovered from scrap.

The use of mercury in the manufacture of certain automobile bumpers began during 1968; therefore, mercury emissions due to disposal during that year were negligible. In future years there may be a serious emission problem.

The remaining 1,239 tons were used in electrical apparatus, instruments, catalysts, and amalgams. Undoubtedly part of the mercury used in this manner will be reclaimed sometime in the future as secondary mercury, and another part will be become waste resulting in substantial atmospheric emissions.

During 1968 the atmospheric emissions due to the disposal

of other mercury were the result of mercury placed in use during previous years. Since these emissions cannot be calculated accurately, all factors have been considered and a reasonable figure of 124 tons has been estimated (10 percent of 1,239 tons) as the mercury emissions to the atmosphere during 1968 due to the disposal of batteries, lamps, instruments, and other items containing mercury.

APPENDIX A

MAJOR MERCURY PRODUCING MINES - 1968 [1]/

State	County	Mine
Properties Producing 1,000 Flasks or More		
California	San Luis Obispo	Buena Vista
California	Marin	Gambonini
California	Santa Barbara	Gibraltar
California	Napa	Knoxville
California	Inyo	Last Chance
California	Sonoma	Mount Jackson
California	San Benito	New Idria
Idaho	Washington	Idaho-Almaden
Nevada	Humboldt	Cordero
Properties Producing 500-1,000 Flasks		
California	Trinity	Altoona
California	Santa Clara	New Almaden
Nevada	Esmeralda	B & B
Nevada	Pershing	Red Bird

1- Minerals Yearbook; Bureau of Mines; 1968.

Properties Producing 100-500 Flasks

Alaska	Aniak	White Mountain
Arizona	Maricopa	National
California	Lake	Abbott
California	Marin	Bueno Chileno
California	Napa	Corona
California	Sonoma	Culver-Baer
California	Santa Clara	Guadalupe
California	Lake	Konocti
California	Kings	Little King
California	San Benito	San Carlos
Nevada	Pershing	Goldbank
Nevada	Pershing	Horton Mercury
Nevada	Washoe	Old West
Oregon	Lane	Black Butte
Oregon	Malheur	Bretz
Oregon	Lake	Glass Butte
Texas	Presidio	Fresno
Texas	Brewster	Study Butte